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A Modified Nitride-Based Fuel for Long Core Life and Proliferation Resistance

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Abstract - *A modified nitride-based uranium fuel to support the small, secured, transportable, and autonomous reactor (SSTAR)[1] concept is initiated at Lawrence Livermore National laboratory (LLNL). This project centers on the evaluation of modified uranium nitride fuels imbedded with other inert (e.g. ZrN), neutron-absorbing (e.g. HfN) , or breeding (e.g. ThN) nitrides to enhance the fuel properties to achieve long core life with a compact reactor design. A long-life fuel could minimize the need for on-site refueling and spent-fuel storage. As a result, it could significantly improve the proliferation resistance of the reactor/fuel systems. This paper discusses the potential benefits and detriments of modified nitride-based fuels using the criteria of compactness, long-life, proliferation resistance, fuel safety, and waste management. Benefits and detriments are then considered in recommending a select set of compositions for further study.*

I. INTRODUCTION

This study describes the research and evaluation of advanced nuclear fuels and associated fuel cycles, and explores the suitability of existing and innovative technologies for addressing the emerging requirements for a small, secure, transportable, and autonomous reactor (SSTAR). The SSTAR concept is targeted for energy markets in locations that are either remote or otherwise lacking large-scale energy infrastructures. Design features related to security, such as sealed and long-life fuels, integral instrumentation and control, and specialized detection and signaling systems will be incorporated to minimize the risk of diversion of nuclear materials.

Uranium mono-nitride fuel was chosen for this study due to favorable properties such as its high actinide density and high thermal conductivity. The thermal conductivity of mono-nitride is 10 times higher than that of oxide (23 $W/m^{-}K$ for UN vs. 2.3 $W/m^{-}K$ for UO_2 at 1000 K)[2] and its melting temperature is much higher than that of metal fuel (2630^{o}C for UN vs. 1132^{o}C for U metal). It also has relatively high actinide density, (13.51 gU/cm^3 in UN vs. 9.66 gU/cm^3 in UO_2) which is essential for a compact core design. Uranium mono-carbide fuels also have favorable properties, but are not as well developed for use in the form of sintered pellets and were not considered for this study.

Uranium mono-nitride fuel is being manufactured at the Lawrence Livermore National laboratory (LLNL) by carbothermic reduction of oxides in a controlled glovebox environment. Details of the fabrication and characterization work are given in a separate paper.

This study examines the properties and performance of uranium mono-nitride fuels and compares them with the properties and performance of various modified nitride-based uranium fuels. For the purpose of this study, modified nitride-based fuels are composed of uranium nitride that is mixed atomically with other inert (*e.g.* ZrN), neutron-absorbing (*e.g.* HfN), or breeding (*e.g.* ThN) nitrides. The logic used in the selection of the modified nitride based fuels is shown schematically in **Figure 1** by the yellow boxes. Once a composition is selected as described in this paper, it will be studied further using fuel performance codes. If the composition selected is still attractive, it will then be fabricated and characterized both before and after irradiation. Data will be compared to UN. Samples will likely be irradiated at the McClellan Nuclear Radiation Center (MNRC).

SSTAR is a small fast reactor controlled by a slow-moving reflector in the vertical direction of the reactor. Key points in the reactor design are that it must have a long core life and a compact design. A long-life fuel (approaching 30 years ideally) could minimize the needs for on-site refueling and spent-fuel storage. As a result, it could significantly improve the proliferation resistance of the reactor/fuel systems. Compact design is needed so that transport of the reactor to remote locations or developing countries is not cost prohibitive. Depending largely upon the energy needs at the location sited, these reactors are envisioned to be in the range of 10KWe to 150MWe.

II. FUEL SELECTION CRITERIA

The primary factors that affect the selection of the reactor fuel are compactness, long-life, proliferation resistance, fuel safety, and waste management. For a modified nitride-based fuel to be selected over pure uranium mono-nitride fuel, it must be superior in several of these factors and comparable in the remaining factors. The focus of the study is on potential benefits of adding group IIIB nitrides (*e.g.* TiN, ZrN, and/or HfN) and other actinide nitrides (ThN and/or PuN) to the base UN fuel.

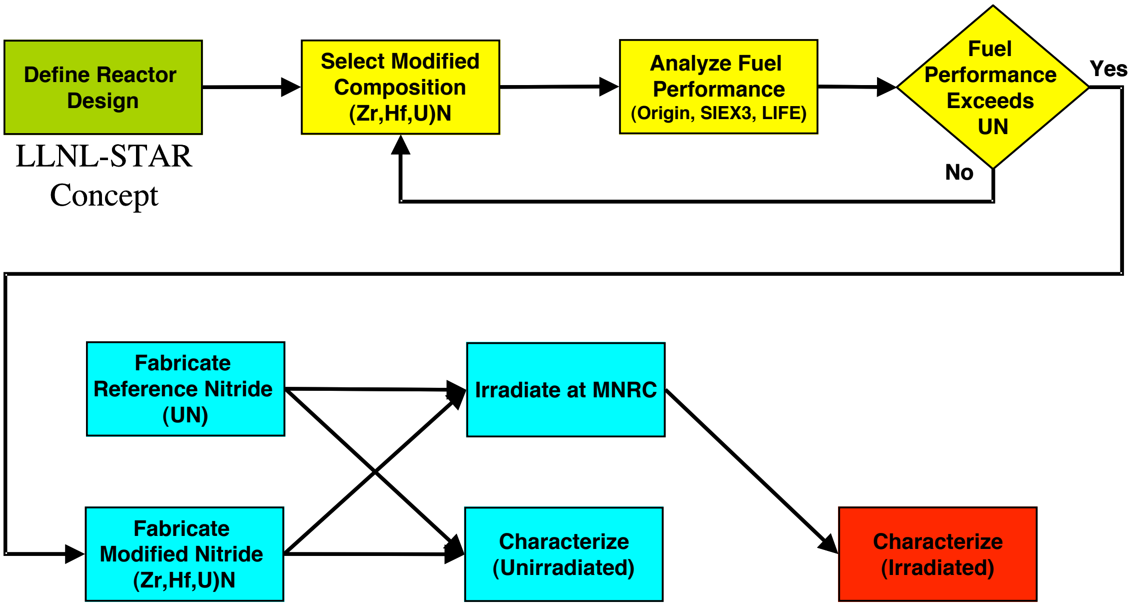

Figure 1. Depiction of Logic Used to Select, Fabricate ,and Characterize Modified Nitride Fuels. Green-design. Yellow-fuel selection. Blue-fabrication and irradiation. Red-post irradiation examination.

1. Compactness

Higher density of the fissionable isotope (^{235}U) favors a more compact reactor because the critical mass will be smaller. A smaller core will yield more heat per unit area and higher thermal conductivity of the fuel will be required to get the heat out of the system and maintain an acceptable centerline temperature of the fuel.

If composition of the fuel can be modified to increase the thermal conductivity without decreasing the density of the ^{235}U in the fuel or affecting the stability of the fuel, a more compact reactor design is favored. **Table 1** shows the thermal conductivity of a variety of nitrides at various temperatures. The data show that additions of ThN should increase the thermal conductivity substantially. Additions of TiN or ZrN should increase the thermal conductivity slightly, and additions of HfN should decrease the thermal conductivity slightly.

Table 1. Thermal Conductivity of Various Nitrides.

Material	T=20°C (W/m-K)	T=500°C (W/m-K)	T=750°C (W/m-K)	T=1000°C (W/m-K)
TiN[3]	19.2	25.3	25.7	25.9
ZrN[3]	20.5	22.7	22.7	23.2
HfN[3]	21.7	16.3	15.3	15.7
ThN[4]	51.5	47.7	46.0	44.8
UN[2]	14.5	20.6	22.7	24.6
PuN[19]	14.0	12.5	12.0	13.0

With the possible exception of ThN, a modified nitride-based fuel will have minimal positive or negative impact on the compactness of the reactor. If the ^{235}U content in the fuel remains the same, the addition of ThN could be very beneficial to the overall thermal conductivity, thus favoring a more compact design.

2. Long-life

Long -life is obtained largely by the reactor design. But fuel composition can also affect the life favorably or unfavorably. The favorable features are:

- High fissile loading (i.e., high ^{235}U enrichment in UN, or high ^{239}Pu content in (U,Pu)N),
- Presence of ^{232}Th or ^{238}U which are converted during irradiation into ^{233}U-rich uranium and ^{239}Pu-rich plutonium, respectively
- Low cladding strain which is dependent upon the fuel centerline temperature and the radiation effects on the fuel and cladding materials
- Presence of burnable poisons in the fuel,

Additions of inert materials such as TiN and ZrN are a detriment to long life and only serve to dilute the active components of the fuel.

Proliferation concerns will limit the ^{235}U enrichment and cladding strain caused by radiation effects is not known for most of the nitrides under consideration in this study. Therefore, this discussion on long core life centers largely on any potential benefits that can be gained by the use of burnable neutron poisons.

Since the SSTAR will be a fast spectrum reactor with a peak neutron energy between 0.2 and 0.3 MeV, the use of burnable poisons will be considerably less effective than in thermal spectrum reactors. **Table 2** gives the neutron absorption cross sections of various elements at a neutron energy of 0.1 and 0.5 MeV. As can be seen in the table, the neutron absorption cross sections in this energy region are relatively low, even for many elements that are normally identified as good neutron poisons. Of all these elements, Eu is the best neutron poison in this energy regime, and Hf, Sm, Gd, and Dy are all considerably better than average neutron poisons.

Table 2. Neutron Absorption Cross Sections[5]

Element	Isotope	Cross Section at 0.1 MeV (barns)	Cross Section at 0.5 MeV (barns)
Ti	Nat	0.0084	0.0039
Zr	Nat	0.026	0.023
Hf	Nat	0.35	0.17
Nd	Nat	0.077	0.047
Sm	Nat	0.39	0.22
Eu	Nat	1.33	0.50
Gd	Nat	0.46	0.22
Dy	Nat	0.36	0.19
Th	232	0.26	0.18
U	235	0.42	0.17
U	238	0.17	0.11
Pu	239	0.40	0.16

Data taken from the NGATLAS neutron capture data library

The fission cross sections of some of the actinides are given in **Table 3**. The table shows that the fission cross section of ^{235}U and ^{239}Pu are roughly the same as Eu and about a factor of four higher than Hf, Sm, Gd, and Dy. Therefore, the rate of neutron capture by one of the burnable poisons will be a little less but still comparable to the rate of fission of the ^{235}U. Even though the neutron absorption cross sections are relatively low in this energy regime, burnable poisons could still lengthen fuel life significantly.

Table 3. Fission Cross Sections[5]

Element	Isotope	Cross Section at 0.1 MeV (barns)	Cross Section at 0.5 MeV (barns)
Th	232	0	0
U	235	1.6	1.1
U	238	0	0
Np	237	0.018	0.45
Pu	239	1.5	1.6

Data taken from the MCNP library

3. Proliferation Resistance

Proliferation resistance is ensured primarily by the reactor design, but composition of the fuel is also a significant consideration. To ensure that the uranium in the fresh fuel is not attractive for use in nuclear weapons, the ^{235}U enrichment will be limited to 20%. The fresh fuel can be made even less attractive for diversion by the addition of other inert materials that are not readily separated from UN. Addition of inert materials that are harder than UN to dissolve in aqueous solutions enhances the proliferation resistance. Once the material is dissolved in solution, a PUREX-like purification process will provide good separation of the actinindes from the other inert materials.

Reaction or dissolution rates of nitrides are not well characterized. **Table 4** summarizes available data and includes the approximate time required to completely react or dissolve the various nitride powders in water (H_2O), concentrated nitric acid (HNO_3), concentrated hydrochloric acid (HCl), and concentrated sulfuric acid (H_2SO_4). Note that water reacts with but does not dissolve ThN and PuN. Likewise concentrated nitric acid reacts with but does not dissolve TiN. All other data in **Table 4** are for complete dissolution.

Overall, it is not clear that any significant benefit is obtained by adding other nitrides to the UN fuel matrix. Addition of ZrN may make dissolution in concentrated nitric acid more difficult, but it will probably make dissolution in hydrochloric and sulfuric acids easier.

Table 4. Reaction and Dissolution Properties of Powdered Nitrides in Water and Concentrated Acids.

Nitride	H_2O	HNO_3	HCl	H_2SO_4
TiN	No rxn[6]	<60 m[6]	No rxn[6]	~6 d[6]
ZrN	No rxn[6]	~4 h[6]	~60 m[6]	~60 m[6]
HfN	No rxn[6]	~2 h[6]	~60m[6]	~60 m[6]
ThN	~20m[7]	No data	No data	No data
UN	No rxn[8]	~30 m[8]	No rxn[8]	No rxn[8]
PuN	~15m[9]	~90 m[9]	> 90 m & <2 d[9]	~2 d[9]

Temperature is of reaction/dissolution is approximately 95°C.

Proliferation resistance is also a consideration in the spent nuclear fuel, especially if the fuel will be reprocessed. If there is ^{232}Th or ^{238}U in the fresh fuel, the irradiation will lead to ^{233}U-rich or ^{239}Pu-rich weapons useable material in the spent fuel. The attractiveness of the Pu is reduced, but not eliminated, by the presence of higher enrichments of ^{235}U in the fresh fuel. Under irradiation, some of the ^{235}U is eventually converted into to ^{238}Pu. The attractiveness of the plutonium in the irradiated fuel is also reduced, but not eliminated, by longer irradiation times, *e.g.* higher burn-ups. The long irradiation cycle (as supported by the long life of the reactor fuel) will render the spent fuel with a higher percentage of ^{240}Pu and ^{242}Pu.

Overall, the attractiveness of the fresh fuel is reduced very little by additions of other nitrides to the UN fuel, and the attractiveness of the spent fuel is reduced some by minimizing the ^{238}U content relative to the ^{235}U

content as long as the ^{235}U enrichment is kept below 20%.

4. Fuel safety

For nitride fuel and cladding, the relevant criteria for fuel safety are

- Fission gas release and retention
- Fuel pellet cladding interaction
- Radiation swelling effects
- Fuel centerline temperature

In general, the attainable burn-up and thus operating life of nuclear fuels are limited by materials performance issues, which result from changes in the thermal and mechanical properties and dimensional stability of the fuel pellets, cladding and structural materials during neutron irradiation. The development of modified nitride fuel will require improved materials and design approaches in order to reach higher burn-ups.

The effects of fission gas build-up can be mitigated by the fuel pin design. If the density of the fuel is low enough that the porosity is open, usually around 95% of theoretical or less, the fission gases will be able to diffuse out of the fuel and collect in the gaps at the ends of the fuel pins. Modification of the fuel composition is not a benefit or detriment in this respect.

The stability of the fuel with the cladding will be dependent upon the thermodynamic stability, compatibility of the materials, the fuel temperature, and radiation induced swelling effects. Thermodynamic stabilities of various nitrides are summarized in **Table 5**. The melting point and heat of formation are qualitative measures of the relative stability of the nitrides and show qualitatively that all the nitrides listed are more stable than UN and PuN.

Table 5. Melting Points and Oxidation Properties of Powdered Nitrides.

Nitride	MP (°C)	$\Delta_f H^0_{298}$ (kJ/mol)	Oxidation Begins (°C)	Ignition Temp. in Air (°C)
TiN	2945[10]	-337.6[11]	580[13]	>680[13]
ZrN	2960[10]	-365.3[11]	600[13]	>740[13]
HfN	3387[10]	-373.6[12]	650[13]	>810[13]
ThN	2827[10]	-391.2[12]	360[4]	520[4]
UN	2762[18]	-290.8[12]	100[15]	~300[15]
PuN	2469[14]	-299.2[12]	<25[9]	~290 dry[15] ~100 moist[15]

Note that MP for UN and PuN is the temperature at which they decompose into a liquid metal and 1 atm of N_2(g)

Also listed in **Table 5** are the temperatures where oxidation of the nitride powder begins and the temperature at which the powder ignites. For bulk solids, these temperatures will of coarse be much higher. The data show clearly that additions of any of the nitrides except PuN will benefit the oxidation resistance and possibly simplify considerably the handling of the material. The best nitrides to add to the fuel matrix are clearly ZrN and HfN.

In regards to interaction with the cladding some thermodynamic calculations can be performed to see if the nitrides are stable with respect to the cladding materials, which are taken to be a zirconium-rich alloy and niobium-rich alloy. **Table 6** gives the free energy of reaction with zirconium or niobium per mole of nitride. For reaction with zirconium, the equation considered is

$$MN + Zr = ZrN + M \quad (1)$$

And for reaction with niobium, the equation considered is

$$MN + 2Nb = Nb_2N + M \quad (2)$$

where M is Ti, Zr, Hf, Th, U, or Pu. Thermodynamic calculations were performed using FactSage 5.0[16]. The components of stainless steels are not easily nitrided so similar reactions will not occur.

Table 6. Thermodynamic Stability of the Nitrides[16]

Nitride	$\Delta_r G^0_{1273}$ with Zr (kJ/mol)	$\Delta_r G^0_{1273}$ with Nb (kJ/mol)
TiN	-29.7	73.7
ZrN	0	103.4
HfN	12.3	115.7
ThN	18.9	122.3
UN	-56.0	47.4
PuN	-55.4	48.0

The free energy data show that UN is stabilized considerably by the addition of HfN or ThN. Without these additions, zirconium is not a suitable cladding material for uranium nitride fuel. In the case of niobium, there are no reactions with the nitrides. Consequently, no there no particular advantages or disadvantages to a modified nitride fuel when using niobium cladding. In the case of nickel, a common component of stainless steels, it is known that UN will react with nickel to form UNi_5 and U_3N_4[17]. It is not known whether or not similar reactions occur with the other nitrides or if iron or other components in stainless steels will undergo similar reactions. Further study is needed to determine whether or not there are any advantages or disadvantages when using stainless steel cladding with a modified nitride fuel.

In summary, additions of HfN and ThN and to some extent ZrN make the fuel considerably less reactive with the cladding and significantly safer to handle in an air atmosphere.

5. Waste Management

Waste management is a complicated issue with spent nuclear fuel. If the fuel is to be used in a once through cycle and disposed of directly, it will be important that the fuel is stable for thousands of years in an underground repository. If the fuel is to be used in a close cycle, it will be important that the fuel can be easily purified and reused.

For an open once through fuel cycle, the spent fuel will eventually need to be disposed of in an underground repository. Since the components in the fuel will not be reused, it is best from a proliferation point of view to dispose of the fuel directly and not separate the fission products from the actinides thus making them attractive for theft or diversion. Nitrides are more reactive than oxides so the case for direct disposal may be more difficult. As already noted, addition of nitrides except PuN to the UN fuel will significantly stabilize the fuel making it more stable with respect to water and more resistant to oxidation. Thus, it is a significant advantage from the waste management point of view for a direct disposal option to add TiN, ZrN, HfN, or ThN to the UN fuel.

For a closed fuel cycle involving reprocessing and reuse of the nuclear materials, the ease of dissolution and purification is an important factor from a waste management point of view. HfN and ZrN appear to be a little harder to dissolve than UN. Thus additions of these nitrides might complicate the purification and reuse of the spent nuclear fuel. Hard to dissolve materials will generally require more complex processing and more wastes will be generated.

Overall, it is probably an advantage to add ZrN or HfN to the fuel rather than use pure UN. In the near future, there appears to be more support in the U.S. for once through open fuel cycles. In the long-term, as the cost of fuel and waste disposal becomes more expensive, closed cycles will likely become more attractive. In a closed cycle, addition of ZrN or HfN to the fuel may complicate the dissolution and purification process.

III. FUEL COMPOSITION SELECTION

Based on the criteria of compactness, long-life, proliferation resistance, fuel safety, and waste management some attractive candidates for a modified nitride-based fuel can be selected and evaluated. Overall, TiN does not offer any advantages over ZrN as an inert material so it is not considered further in this first phase of study. Due to current limitations in the LLNL nitride fabrication glove box, additions of ThN and PuN are not considered for the moment. This leaves four parameters: enrichment, UN content, ZrN content, and HfN content.

Table 7 summarizes the four parameters against the five selection criteria. A plus means that the parameter is a slight benefit for that criteria and a minus means that that parameter is a slight detriment to that factor. A zero indicates little or no effect. A double plus indicates a large beneficial effect and a double minus indicates a large detrimental effect.

Table 7. Summary Fuel Selection Criteria

Metric	**Enrich.**	**UN**	**ZrN**	**HfN**
Compactness	+	0	+	-
Long-Life	++	+	--	+
Prolif. Res.	--	-	+	+
Fuel Safety	0	--	++	++
Waste Man.	0	-	+	+

In summary, the ^{235}U enrichment should be as high as possible with out exceeding 20%. HfN should be added in a suitable amount as a burnable poison, and the balance of the fuel should be ZrN.

Given these criteria, four compositions are suggested for further study in **Table 8**. Case 1 is pure UN with maximum allowable enrichment of ^{235}U. Case 2 is the reference case of UN with 10% enrichment of ^{235}U. Case 3 is an intermediate case with a limited amount of ZrN and HfN added. Ratio of Hf-to-^{235}U is chosen to be 1-to-1. Case 4 is the limiting case where the maximum amount of ZrN is added while limiting the ^{235}U enrichment to 20%. Again the ratio of Hf-to-^{235}U is chosen to be 1-to-1.

Table 8. Selected Compositions for Further Study

	^{235}UN	**^{238}UN**	**ZrN**	**HfN**
Case 1	20	80	0	0
Case 2	10	90	0	0
Case 3	10	70	10	10
Case 4	10	40	40	10

Compositions are given in an atomic or molar basis.

In future studies, additions of ThN and/or PuN may be considered for further study. In terms of compactness and fuel life there could be a significant benefit by replacing ZrN with ^{232}ThN, which has a higher thermal conductivity and breeds during irradiation to ^{233}U. If proliferation concerns in the freshly fabricated fuel can be address by other means, ^{235}U can be replaced by ^{239}Pu in the freshly fabricated fuel.

IV. FUTURE FUEL ANALYSIS

The thermal and mechanical performance of the modified nitride fuels with compositions listed on **Table**

7 will be modeled and evaluated by computer codes SPACEPIN, SIEX3 and LIFE4Rev1. The SPACEPIN is a code used to model the UN fuel for SP-100. The fuel was cladded in Nb-1Zr tubing lined with Re. For this study, a common stainless steel may be used as cladding. SIEX3 and LIFE4Rev1 are computer codes designed for advanced fuel for liquid-metal fast reactors. They will be used to model the modified nitride fuel with different cladding materials including HT-9, 304 SS, 316 SS, and In 706. The fuel performance analysis will determine the history of the fuel centerline temperature, the volume changes by cracking, restructuring, densification and swelling, the amount of fission gas release and retention, as well as the degree of cladding strain during the irradiation period.

For neutronic calculation, computational nuclear design tools, such as MOCUP will be used to examine the neutron absorption characteristics of the fuel and claddings materials. MOCUP is a package of computer codes coupling MCNP and ORIGEN2, and can be used to simulate the depletion of general materials (fuel, target, cladding, coolant, control rods, etc.) in a fast-spectrum field.

V. CONCLUSION

A study of candidate materials for a modified nitride-based fuel has been completed. The candidate fuels for further study are selected by the potential benefits based on five criteria: compactness, long-life, proliferation resistance, fuel safety, and waste management. The nitrides considered are TiN, ZrN, HfN and ThN with the base nitride fuel being UN or possibly (U,Pu)N.

Overall, there are potentially significant advantages to using modified nitride based fuels. Compositions high in ZrN (or ThN) with a small amount of HfN are recommended for further study. These compositions will be evaluated by various computer codes and they will be fabricated, irradiated, and characterized both before and after irradiation following the logic presented in **Figure 1**.

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